STUDIES ON RADIONUCLIDE DISTRIBUTION AND RADON EXHALATION RATE IN THE ENVIRONS OF DAVANAGERE DISTRICT OF SOUTH INDIA

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Submitted by

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Introduction

The phenomenon of radioactivity was discovered by Henry Becquerel in the year 1896. Later, it was found that there are some elements in the environment which exhibit radioactivity and these elements are present in earth's crust ever since its formation. It was soon realized that in the earth crust these radioactive elements are not uniformly distributed. The discovery of artificial radioactivity has added a new era to the studies on radioactivity and ionizing radiation.

The assessment of gamma radiation dose to the human population through natural background radiation is important because natural radiations are the largest contributors to the population dose around the world. Human beings are continuously exposed to ionizing radiation that exists outside and within the planet (UNSCEAR, 2000). The two important sources of natural radiation exposures are terrestrial sources and extra – terrestrial sources. The terrestrial radiation originates from the radionuclides present in the earth crust and the extraterrestrial radiation comes from cosmic rays that penetrate the earth's atmosphere from the outer space. The total radiation dose to human inhabitants is around 2.8 mSv per year, out of which 2.4 mSv of total radiation dose come from the natural sources (Hendry, et al. 2009).

The natural radioactivity in the environment originate from natural radionuclides which are part of the radioactive series such as ^{238}U, ^{232}Th, and singly occurring radionuclide ^{40}K in different geological and geochemical formations. Since, about 98% of gamma dose is received from ^{226}Ra and its progeny, the daughter radionuclides of ^{226}Ra in ^{238}U series are

ignored (UNSCEAR, 2000). People have always been exposed to ionizing radiation from the Naturally Occurring Radioactive Materials (NORM). In the modern world, the exposure to ionizing radiation has increased due to the development in science and technology with the increasing number of heavy radioactivity installations and nuclear activity. The sources of artificial radioactivity are due to multiplication of nuclear power reactor, open air nuclear weapon test, radioisotope usage in the field of nuclear medicine, agriculture and industry. In nuclear reactors, large numbers of radionuclide are released to the environment, which make use in the fission process.

In addition to the above, the production and use of phosphate fertilizer and the electrical power generation through coal based thermal power station has increased the natural radiation level in the environment (UNSCEAR, 2016). These human activities have enhanced the level of radiation and caused the undesirable effects on the living organisms. In recent years there have been growing interest among public, regarding people to ionizing radiation and many researchers around the world have collected data of the public exposure to various ionizing radiation.

The ^{222}Rn is radioactive gas having a half life of 3.82 days originates from the decay of ^{226}Ra, a member in the uranium series (^{238}U). People spend almost 80% of their life inside the house or offices so it is important to find the radiation exposure to the population due to indoor radon. ^{226}Ra forms ^{222}Rn through alpha decay. The radon atoms get migrated through diffusion or convective effect. These two parameters are affected by many other parameters such as uranium concentration, radium concentration, soil texture, permeability and grain size (Ishimori et al., 2013).

Radon is a radioactive gas which emits alpha particles with 5.48 MeV energy. The three natural isotopes of radon are ^{219}Rn (action) decays from the ^{235}U series, ^{220}Rn (thoron)

decays from the ^{238}U series. Since ^{235}U activity is less abundant and the half-life of ^{219}Rn is 3.96 s, the radiation exposure is not significant for humans. ^{220}Rn with half-life 55.6 s and ^{222}Rn with half-life 3.84 days are of major concern, in terms of radiation exposure to humans (BEIRRes, 1999).

The radon exhaled from soil surface per unit area per unit time, to the atmosphere is known as radon exhalation rate. The radon emanation is one of the important phenomenon which influence the outdoor and indoor radon concentration. In the emanation process, radon escapes from the grains mainly because of recoil in to interatomic space between grains. After the emanation process transportation take place. In the transportation process, emanated radon moves through the residue surface due to the advective flow and diffusion flow. Finally in the exhalation process, the radon which is present in the ground surface are then exhaled to the outer atmosphere. Radon is a noble gas which is exhaled rapidly after breathed in, radon and its daughter products combine with other molecules in air and with dust particles and aerosols or smoke. These particles deposit in the airways of lungs, since radon progeny emits an ionizing radiation as alpha particles. After deposit in the lungs, it can damage cells lining and experiments have been confirmed that ionizing radiation affects the bronchial epithelial and could cause lung cancer (Menon et al., 2015).

The radionuclide ^{210}Pb is another member of ^{238}U series and it occurs in nature in partial radioactive equilibrium with its daughters ^{210}Bi and ^{210}Po. The complete equilibrium of ^{210}Pb - ^{210}Bi - ^{210}Po in the biosphere is usually not reached due to the difference in the influence of biological, meteorological, chemical and other factors on each of these radionuclides (Jaworowski, 1967). Lead-210, which has a relatively long half-life of 22 years decays to ^{210}Bi through beta emission. The short lived ^{210}Bi ($T_{1/2}$= 5 days) decays by beta emission forming ^{210}Po. Both ^{210}Pb and ^{210}Po easily enter into the human body by the way

Consumption of food is usually the most important route to enter the human organs. The radionuclide ^{210}Pb is a bone seeker and is found incorporated in bone mineral from where it seems to be eliminated by long term skeletal remodelling (Lloyd et al., 1974). Polonium-210 in contrast is not a bone seeker but is rather distributed in soft tissues after intake. Polonium-210 decays with a half-life of 138 days by alpha emission (5.3 MeV). Therefore, ^{210}Po causes considerably greater biological damage compared to beta emitter ^{210}Pb. It is estimated that the equivalent dose resulting from a single disintegration of ^{210}Po is a thousand times greater than ^{210}Pb decay (Parfenov, 1974; Morgan et al., 1964). Hence, ^{210}Po is included in the group of most toxic radioisotopes.

Analysis of the radionuclide continent of water, soil, rock, food, air and knowledge of the metabolic behaviour of investigations are needed to obtain a clear understanding of the basic dynamics of radioactivity in the environment. The external gamma radiation exposure to the population changes due to the geology and geographical condition of the area and its associated radioactivity level in various parts of the soil (Linsalata, 1994). Many researchers have studied the radiation level in various parts of the word has been reported and the distribution of natural radionuclides in environmental matrices such as rocks and soil (Papp et., 2002; Sivakumar et al., 2012; Ozmen et al., 2014; Ozden et al., 2013; Rangaswamy et al., 2015; Prakash et al., 2017; Michel et al., 2019). A more extensive and systematic investigations are needed to obtain a clear understanding of the basic dynamics of radioactivity in the environment.

Literature shows that, the measurement of radionuclides activity in soil and rock are very much important because of these are the major source of radionuclides activity. So an extensive and systematic investigation is necessary to understand the dynamics of radionuclides in the environment. Therefore, in the present study the Davangere district has

been taken to carried out the radioactivity concentration measurement. Davanagere district occupies an area of 5926 km^2, which has six taluk viz., Channagiri, Honnali, Harihara, Harapanahalli, Jagalur and Davanagere. The minerals are well concentrated in this region such as manganese ores, iron ores, quartz and white quartz. The important rocks available in this district are metamorphic rocks, plutonic rocks, volcanic/metavolcanic, gneiss and quartz. (District mineral survey report 2008). The district is present in the mid – eastern region of the Karnataka state between 13º 45'00" N and 14º 50' 00" N latitude and 75º 30' 00" E and 76º 30' 00" E longitude. To understand the radionuclide dynamics and dose received from soil and rock samples in Davanagere district of Karnataka in India, systematic investigations were carried out and reported in this Thesis.

Aim and Scope

Davanagere district one of the mineral rich district in Karnataka, India.The region consists of varieties of rocks as well as minerals. The minerals found in this region are manganese ores, iron ores, quartz and white quartz. The important rocks available in this district are metamorphic rocks, plutonic rocks, volcanic/metavolcanic, gneiss and quartz. The district is placed under major sedimentary components called the younger schist belt of Dharwar type. This belt from the main extent of the schistose rocks with the exposed island of basement gneissic complex at Honnalli region the youngest formation of an acid volcanic suit is well exposed in Malebennur of Harihara Taluk in the region of Davanagere district.

Mining is one of the parameter of anthropogenic activities and negatively impact the environment by causing destruction. The ore mining and processing in environment are the source of trace metals. It will take several decades from the recovery of ecosystem from mining activities. As a result, physico-chemical properties of soil changes and it create other environmental problems. Hence studies on radionuclides in soil, radon exhalation rate,

polonium and lead activities. The present study aims to find the activity concentration of radionuclides due to the clay minerals and radioactive elements in the region.

In view of this, systematic investigations have been carried out on the distribution of radionuclides dependence upon the clay minerals in the Davanagere district of southern India. The study includes the estimation of activity concentration of natural radionuclides like ^{226}Ra, ^{232}Th, and ^{40}K in soil and rock samples using NaI(TI) spectrometer. The study of seasonal variation of natural radionuclides in soil is important, to study the transport of radionuclides from rock to soil phase. The assessment of radiation hazard parameters from the obtained natural radionuclide activity is important, which gives information on the internal and external exposure to the people in the region.

The measurement of gamma radiation dose from the natural sources is particularly important, because of it is the largest contributor to the total exposure of population. Therefore, in the present investigation, the gamma dose rate in air was measured along the sampling location with a gamma ray survey meter. The radon exhalation rate depends upon several parameters like radium concentration, soil texture, permeability. The radon exhalation in soil samples were assessed using sealed can technique. The assessment of radiological hazards is important, keeping in view of health hazard effects of radon in the environment. The activity concentration of ^{210}Po and ^{210}Pb in soil samples were analyzed using radiochemical deposition method.

The variation in activity concentration of radionuclides may be contributed by several factors in the time of formation of soil and rock. Therefore, the effect of physico-chemical properties of radio elements distribution has been examined by using standard protocols. This play a major role in determining the behaviour of radionuclides present in it. In the present study, the clay minerals were identified by using the FTIR spectrometry.

The research study under taken and presented in this thesis enrich the global data bank on natural radioactivity which are potentially important for the purpose of radiation risk assessment. The results discussed in the thesis could help to minimise the health risk associated with it by adopting preventive measures wherever felt necessary.

Methodology

- In the present study, the energy compensated GM tube based Polimaster (PM1405) gamma ray survey meter was used to measure the ambient gamma dose rate in the air.
- The sampling locations were identified based on geology of individual locations. The soil and rock sampling were collected along the mining area system, following the EML standard procedures (EML, 1983).
- The samples were collected during pre monsoon and post monsoon. The collected samples were brought to laboratory for further processing. The samples were dried in air circulated oven and sieved through 250 micron mesh. The sieved samples were stored in 250 ml container to obtain secular equilibrium between radium-226 and its progeny
- In the present study, NaI(TI) gamma spectrometry system was used to determine the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the rock and soil samples. The spectrometer efficiency and energy was calibrated using the IAEA standard sources.
- The concentration of alpha active radionuclide ^{210}Po was estimated by radiochemical separation and subsequent deposition on a silver disc. The disc was then counted on both sides using a ZnS(Ag) alpha counter having 30% efficiency. The activity of ^{210}Pb was estimated through ^{210}Po by allowing the ^{210}Po plated solution for a period of 12 months to build up the ^{210}Po from ^{210}Pb.

- The FTIR spectrometry was used in the present study to analyse the mineralogical composition of the soil and rock samples in the environment. KBr pellet technique was employed in preparing the sample for FTIR spectrometry (Ramasamy et al., 2014) and the minerals were identified by comparing the transmittance spectra of the samples with standard values.
- Multivariate statistical analysis was used to find the interrelation among the parameters obtained from the rock and soil samples. In the present investigation, the statistical software 'Statistical Program for the Social Science (SPSS) statistical analysis and SURFER software was used for the 3D map.

The Thesis

The thesis is divided into five chapters.

Chapter 1 constitutes an introduction to the studies undertaken in the present work. Sources of natural and artificial radioactivity in the environment, details of all natural radionuclides, the relevance of the study and a detailed survey of the literature providing a glimpse of the work done in this area by various investigators all over the world are outlined. The description of the study area, aim and scope of the present study are also presented at the end of this chapter.

Chapter 2 deals with the details of experimental techniques followed and the instruments used in the present investigation. Details of the instruments like gamma survey meter, alpha counting system, NaI(TI) gamma ray spectrometry, LR – 115 (type II) detector, spark counter, and FTIR spectroscopy were used to carry out the present work and their salient features are discussed in this chapter.

Chapter 3 provides a detailed discussion on selection of the sampling stations, collection of samples, sample processing and radiochemical methods followed to separate

various radionuclides. The methods employed to calculate the activities concentration of radionuclides, radon exhalation rate in the environmental samples from measured counts and estimation of radiation hazard to the population of the region are discussed in this chapter.

The results of the present study are discussed in the Chapter 4. This result includes gamma absorbed dose rates in air, the distribution and enrichment of prominent natural radionuclides in soil and rock, radon exhalation rate in the soil, calculated radiation hazard parameters are compared with recommended limit to assess the radiation exposure to the human population in the study area. This chapter also includes the dependence of physico – chemical parameters of soil with statistical analysis. The relation between the natural radionuclides and minerals are also included.

Chapter 5 summarises important findings of the present study. The suggestions for future study in the related fields are discussed at the end of this chapter.

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Research Guide Research Student
Date:

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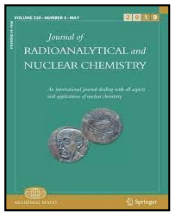
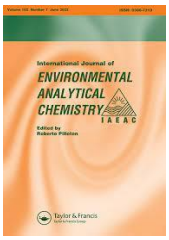
LIST OF PUBLICATIONS

Research articles published in Journals (Peer-Reviewed)

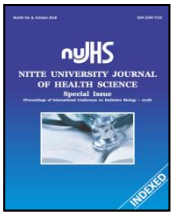
Part of the Thesis

[1] 

Malleshi Kavasara., Vinutha, P.R., Kaliprasad, C.S., & Narayana, Y. (2021). Studies on the dependence of natural radioactivity on clay minerals of soils in Davanagere district of Karnataka, India. *Journal of Radioanalytical and Nuclear Chemistry*, 330 (3), 1461–1471.

[2] 

Malleshi Kavasara., Vinutha, P.R., Kaliprasad, C.S., & Narayana, Y. **(**2022**).** Assessment of effective dose and radiological risk from natural radioactivity in rock samples of Davanagere district, Karnataka, India. *International Journal of Environmental Analytical Chemistry*, ISSN: 1029-039.

[3] 

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Malleshi Kavasara., Vinutha, P.R., Kaliprasad, C.S, Prakash M.M., & Narayana, Y. (2022). Speciation and Behavior of ^{210}Po and ^{210}Pb activity in soil around the mining activity region of Davanagere district, Karnataka in India. *Radiation Protection Dosimetry*, ISSN 1742-3406 (Communicated).

Other related publications

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[1] **Malleshi Kavasara.,** Narayana Y., Kaliprasad C. S., Surabhi P. P, Snehalatha T.H., & Vinutha P R., Distribution of radionuclides in medicinal plants of Uttar Kannada district in costal Karnataka, *International conference on radiation biology (ICRB-2018)*, K S Hegde medical college, Kshema Campus, Mangalore. October 4-6, 2018.

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[3] **Malleshi Kavasara.,** Vinuth P.R., Kaliprasad C.S., & Narayana Y., Distribution of natural radionuclides in the soils and assessment of radiation hazards in the Davanagere district of Karnataka, India. *21st National Conference on Solid State Nuclear Track Detectors and Their Applications (SSNTDs-21).* New Delhi, January 29-31, 2021.

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[5] **Malleshi Kavasara.,** participated in the one day workshop on Scientific data analysis using origin pro and mathematical organized by the department of post graduate and research in physics, Bangalore University, held on 2nd January 2020.